Orbital Magnetization in Extended Systems

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While the orbital magnetic dipole moment of any finite sample is well-defined, it becomes ill-defined in the thermodynamic limit as a result of the unboundedness of the position operator. Effects due to surface currents and to bulk magnetization are not easily disentangled. The corresponding electrical problem, where surface charges and bulk polarization appear as entangled, was solved about a decade ago by the modern theory of polarization, based on a Berry phase. We follow a similar path here, making progress toward a bulk expression for the orbital magnetization in an insulator represented by a lattice-periodic Hamiltonian with broken time-reversal symmetry. We therefore limit ourselves to the case where the macroscopic (i.e. cell-averaged) magnetic field vanishes. We derive an expression for the contribution to the magnetization arising from the circulating currents internal to the bulk Wannier functions, and then transform to obtain a Brillouin zone integral involving the occupied Bloch orbitals. A version suitable for practical implementation in discretized reciprocal space is also derived, and the gauge invariance of both versions is explicitly shown. However, tests on a tight-binding model indicate the presence of additional edge currents, and it remains to be determined whether these can be related to the bulk band structure.

1. Introduction

In light of growing recent interest in issues of charge and spin transport in magnetic materials and nanostructures, it is an anomaly that there is no accepted expression for the orbital magnetization of an insulator with broken time-reversal symmetry. The analogous problem for the electrical polarization was solved about a decade ago by the modern theory of polarization, based on a Berry phase.[1,2] A similar theory for the case of orbital magnetization is still lacking and is sorely needed. We present here the first step towards such a theory.

The magnetization **M** of a condensed, macroscopically homogeneous, sample is an intensive quantity formally defined as the magnetic dipole **m** per unit volume. Phenomenologically, this is expected to be a bulk property. In this work, we focus on the term in **m** due to purely orbital currents; for a finite sample it is defined as Equation (1):

$$\mathbf{m} = \frac{1}{2c}\int \mathrm{d}\mathbf{r}\, \mathbf{r} \times \mathbf{j}(\mathbf{r}) \qquad (1)$$

However, due to the presence of the position operator **r** in Equation (1), a current circulating on the boundary of the sample affects the value of **m** in an extensive way, thus causing the bulk magnetization **M** to apparently depend on surface, as opposed to bulk, properties of the sample.

So far, there have been basically two approaches to addressing the magnetization of a condensed sample: 1) linear response theory, and 2) decomposition of **j**(**r**) into local ring currents. In case 1, one addresses by definition only the magnetization linearly induced by a given source. It has been shown in 1996 that such magnetization can be expressed in a boundary-insensitive form, implementable in electronic–structure calculations under periodic boundary conditions.[3] This was achieved in a reciprocal-space framework, where the **r**-operator drawback is circumvented by performing a long-wavelength limit. More recently, Sebastiani and Parrinello[4] proposed a different linear-response approach, formulated in real space, and where the **r** operator is tamed by using localized Wannier orbitals. This second flavor of linear response is aimed at (and implemented for) large supercells, where well-separated localized orbitals can be constructed. Both approaches have been used successfully to address several magnetization-related properties of bulk materials.[5,6]

In case 2, the aim is to address and compute **M** as a ground-state bulk property of the condensed material (whenever a nonzero value is allowed by the symmetry of the Hamiltonian), with no reference to any kind of perturbation. In the existing literature[7,8] this is achieved at the price of introducing an arbitrary atomiclike (or cellular) decomposition of the microscopic current **j**(**r**) into localized ring currents. Unfortunately, different decompositions may well lead to very different computed **M** values, similarly to what is very well known to happen for the analogous electrical case.

The theory we present here concerns a contribution $\mathbf{M}_0$ to the magnetization that can be defined as a ground-state bulk property of a crystalline insulator in vanishing macroscopic magnetic field by means of a very different kind of decomposition, based on Wannier functions (WFs). Eventually, the formulation is equivalently recast as a Brillouin zone (BZ) integral. There are two main differences with respect to previous work which are worth emphasizing. First, despite the fact that a WF

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decomposition suffers from a large arbitrariness, the resulting value of $\mathbf{M}_0$ is unambiguous. Second, our $\mathbf{M}_0$ value is obtained from a decomposition of the one-particle density matrix ρ and *not* of the current $\mathbf{j}(\mathbf{r})$ itself. In fact, a knowledge of the bulk $\mathbf{j}(\mathbf{r})$ is insufficient in principle to determine $\mathbf{M}_0$, just as a knowledge of the bulk charge density $n(\mathbf{r})$ is insufficient in principle to determine the polarization. Thus, both features have a close analogy with the corresponding electrical case.[1,2]

The restrictions to the case of insulators in zero macroscopic magnetic field are significant ones. While the restriction to insulators is essential for the theory of electric polarization, we suspect that it is inessential for the case of orbital magnetization, and that future generalizations to metals may not be difficult. On the other hand, the restriction to vanishing macroscopic magnetic field is required in order for the Bloch wave vectors to be good quantum numbers, and the lifting of this restriction may be more difficult. Nevertheless, we regard our work as a first step in the direction of a more general theory. Even with these restrictions, it is important, as a matter of principle, to understand whether the Berry phase theory of electric polarization has analogues in the theory of magnetization. Furthermore, the magnetization properties are closely connected to the theory of linear response to a small applied magnetic field. For example, the magnetization is itself trivially related to the derivative of the total energy with respect to applied field. Furthermore, a theory of orbital magnetization in zero field would immediately provide an alternative route to the calculation of NMR shielding tensors, which can be defined in terms of the local field at nuclear site $\mathbf{R}_s$ linearly induced by the local macroscopic magnetic field.[9] However, by a thermodynamic relation (i.e. equality of mixed partials), they can alternatively be defined in terms of the macroscopic magnetization $\mathbf{M}$ linearly induced by the perturbation generated by a classical pointlike magnetic dipole μ_s at nuclear site $\mathbf{R}_s$ (and its lattice-periodic replicas). This periodic field averages to zero over the cell and its macroscopic value therefore vanishes. (The entire situation is analogous to the difficulty of computing the Born dynamical charge tensor in terms of the force appearing on an atom in a first-order applied electric field; this difficulty can be overcome by computing it alternatively as the derivative of the induced polarization $\mathbf{P}$ with respect to a sublattice displacement in zero macroscopic electric field.) Once a robust formulation is in hand for computing the orbital magnetization, the latter approach should allow for straightforward computations of the chemical shielding tensors by finite-difference methods. Thus, we believe there are ample motivations for developing such a formulation.

In Section 2, we define the problem and introduce our formalism. In Section 3, we derive our reciprocal-space expression for the bulk magnetization of a crystalline insulator, Equation (14), which is the principal result of this work. In Section 4, we show how Equation (14) can be implemented in practical calculations by discretizing it in a numerically gauge-invariant way over a reciprocal-space grid. Section 5 presents some tests in the context of a tight-binding model, confirming the expression for the part of the magnetization related to the circulation of a bulk WF, but indicating the presence of additional edge currents whose nature remains to be full clarified. Finally, in Section 6 we draw some brief conclusions.

2. Generalities

We work at the Kohn–Sham (KS) level, implicitly using density-only functionals[10] while disregarding subtle issues related to current–density functionals.[11] We assume a lattice-periodical, though time-reversal breaking, KS Hamiltonian H. In principle, such breaking could be due to the presence of a staggered magnetic field that averages to zero over the unit cell, so that the vector potential $\mathbf{A}(\mathbf{r})$ can be chosen as lattice-periodic, or to the presence of a large magnetic field that introduces an integral number of flux quanta per unit cell. Neither of these situations arises practically.

However, it can also arise, for example, from the spin–orbit coupling of the electrons of interest (say, itinerant sd electrons) to a background of ordered spins (say, on rare-earth ions) in certain classes of crystals.[12–16] In any case, we limit ourselves to addressing the macroscopic magnetization in a vanishing macroscopic magnetic field. The context of the present theory is therefore completely analogous to that of the modern theory of polarization which, in its standard formulation, addresses the dielectric polarization of a crystalline sample in a vanishing macroscopic electric field.[1,2]

We treat here one isolated spin channel; the case of degenerate spins is trivially obtained by inserting factors of two, while the case of general mixed spins under spin–orbit interaction must be treated with somewhat more care. We use atomic units throughout. The translational invariance of H implies the usual Bloch form for the KS orbitals. For the sake of simplicity, the magnetic gauge, hence the Hamiltonian, is kept *fixed*, although relaxing this condition is not difficult. The remaining gauge freedom, discussed in the following, only concerns the phases of the Bloch orbitals [Eq. (2)]:

$$\psi_{n,\mathbf{q}}(\mathbf{r}) = e^{i\mathbf{q}\cdot\mathbf{r}} u_{n,\mathbf{q}}(\mathbf{r}) \tag{2}$$

at different wavevectors $\mathbf{q}$.

We also exclude from our considerations the case of so-called crystalline Hall insulators.[12,13] These are insulators that display a nonzero dissipationless transverse conductivity σ_{xy} in a fashion completely analogous to the behavior of a filled Landau level in the quantum Hall effect. Such insulators are characterized by an integer topological invariant known as the Chern number (see discussion following Equation (16) in Section 3); indeed, σ_{xy} is just $-e^2/h$ times the Chern number.[17] Because it is not possible to choose a smooth periodic gauge [Eq. (3)]:

$$\psi_{n,\mathbf{q}+\mathbf{G}}(\mathbf{r}) = \psi_{n,\mathbf{q}}(\mathbf{r}) \tag{3}$$

(where $\mathbf{G}$ is any reciprocal lattice vector) for such systems,[12] it is not clear that a Wannier representation is possible. Thus, we exclude this case, and treat only conventional insulators for which the periodic gauge condition, Equation (3), is satisfied.

The Schrödinger equation for the orbitals u is Equation (4),

$$H(\mathbf{q})|u_{n,\mathbf{q}}\rangle = \varepsilon(\mathbf{q})|u_{n,\mathbf{q}}\rangle \tag{4}$$

where $H(\mathbf{q})$ is given by Equation (5):

$$H(\mathbf{q}) = e^{-i\mathbf{q}\cdot\mathbf{r}} H e^{i\mathbf{q}\cdot\mathbf{r}} \tag{5}$$

The velocity operator is defined via Equation (6):

$$\mathbf{v} = i[H,\mathbf{r}] \tag{6}$$

Note that $\mathbf{v}$ is generally different from the canonical momentum $\mathbf{p}$; for example, if $H=\frac{1}{2}(\mathbf{p}+\frac{1}{c}\mathbf{A})^2+V(\mathbf{r})$, then $\mathbf{v}=\mathbf{p}+\frac{1}{c}\mathbf{A}$ and $H(\mathbf{q})=\frac{1}{2}(\mathbf{v}+\mathbf{q})^2+V(\mathbf{r})$. In the case of tight-binding models, one normally does not bother to define $\mathbf{p}$; one just uses $\mathbf{v}$ defined by Equation (6).

For an insulating system with n_b bands, the single-particle density matrix is Equation (7),

$$\rho(\mathbf{r},\mathbf{r}') = \frac{\Omega}{(2\pi)^3} \sum_{n=1}^{n_b} \int_{\mathrm{BZ}} \mathrm{d}\mathbf{q}\, \psi_{n,\mathbf{q}}(\mathbf{r}) \psi^*_{n,\mathbf{q}}(\mathbf{r}') \tag{7}$$

where BZ is the Brillouin zone and the Bloch functions are normalized to the unit cell of volume Ω. For any finite system, the magnetic dipole moment is easily expressed in terms of ρ. However, the expression involves the operator $\mathbf{r}\times\mathbf{v}$, and therefore becomes ill-defined when we try to apply the same expression to an extended system using Equation (7). The analogous problem for the electric dipole can be solved by expressing ρ in terms of WFs; we are going to follow a similar path here.

3. Formalism in Terms of Wannier Functions

The WFs are defined via Equation (8):

$$w_n(\mathbf{r}) = \frac{\Omega}{(2\pi)^3} \int_{\mathrm{BZ}} \mathrm{d}\mathbf{q} e^{i\mathbf{q}\cdot\mathbf{r}} u_{n,\mathbf{q}}(\mathbf{r}), \tag{8}$$

and the density matrix ρ is identically rewritten as Equation (9),

$$\rho(\mathbf{r},\mathbf{r}') \sum_{n=1}^{n_b} \sum_{l} w_n(\mathbf{r}-\mathbf{R}_l) w_n^*(\mathbf{r}'-\mathbf{R}_l), \tag{9}$$

where $\mathbf{R}_l$ are lattice vectors.

While the individual WFs are gauge-dependent, the density matrix is a gauge-invariant and lattice-periodical operator. Written in the form of Equation (9), ρ projects over a set of well localized orbitals (the WFs) having, for example, a finite second spherical moment.[18] The $\mathbf{r}\times\mathbf{v}$ operator then becomes harmless, and the magnetic dipole moment corresponding to each term in the sums in Equation (9) can be evaluated in the usual way. Intuitively, each WF carries in general an electronic current which circulates in a limited region of space. In view of the translational symmetry, we define a contribution $\mathbf{M}_0$ to the macroscopic magnetization of the crystalline sample as the sum of the magnetic dipole moments of the WFs in a cell divided by the cell volume Ω [Eq. (10)]:

$$\mathbf{M}_0 = -\frac{1}{2c\Omega} \sum_{n=1}^{n_b} \langle w_n|\mathbf{r}\times\mathbf{v}|w_n\rangle \tag{10}$$

Since the macroscopic current carried by each band vanishes, it follows that $\langle w_n|\mathbf{v}|w_n\rangle = 0$, so that the choice of origin in Equation (10) is irrelevant. We thus expect $\langle w_n|\mathbf{r}\times\mathbf{v}|w_n\rangle$ to be a fully gauge-invariant quantity. In this respect we are making an even stronger statement than in the electrical analogy. There, the origin independence of the polarization is restored only when the ionic contribution is added to the electronic one, and even then $\langle w_n|\mathbf{r}|w_n\rangle$ is only gauge-invariant modulo a lattice constant (the "quantum of polarization"). In contrast, there is no corresponding "quantum of magnetization" to worry about here.

We manipulate Equation (10) so that it is expressed in terms of BZ integrals of the Bloch orbitals, and prove its gauge invariance, as follows. First, we can write Equation (10) as Equation (11),

$$\begin{aligned} M_{0,\alpha} &= -\frac{\varepsilon_{\alpha\beta\gamma}}{2c\Omega} \sum_{n=1}^{n_b} \langle w_n|r_\beta v_\gamma|w_n\rangle \\ &= -\frac{i\varepsilon_{\alpha\beta\gamma}}{2c\Omega} \sum_{n=1}^{n_b} \langle w_n|r_\beta [H,r_\gamma]|w_n\rangle \\ &= -\frac{i\varepsilon_{\alpha\beta\gamma}}{2c\Omega} \sum_{n=1}^{n_b} \langle w_n|r_\beta H r_\gamma|w_n\rangle \end{aligned} \tag{11}$$

since $\varepsilon_{\alpha\beta\gamma} r_\beta r_\gamma = 0$. Next, a straightforward manipulation[19] of Equation (8) gives Equation (12):

$$r_\gamma|w_n\rangle = \frac{i\Omega}{(2\pi)^3} \int_{\mathrm{BZ}} \mathrm{d}\mathbf{q} e^{i\mathbf{q}\cdot\mathbf{r}} \left|\frac{\partial u_{n,\mathbf{q}}}{\partial q_\gamma}\right\rangle \tag{12}$$

Substituting this into Equation (11) yields Equation (13):

$$M_{0,\alpha} = -\frac{i\varepsilon_{\alpha\beta\gamma}\Omega}{2c(2\pi)^6} \sum_n \int_{\mathrm{BZ}} \mathrm{d}\mathbf{q} \int_{\mathrm{BZ}} \mathrm{d}\mathbf{q}' \left\langle \frac{\partial u_{n,\mathbf{q}'}}{\partial \mathrm{q}'_\beta} \middle| e^{-i\mathbf{q}'\cdot\mathbf{r}} H e^{i\mathbf{q}\cdot\mathbf{r}} \middle| \frac{\partial u_{n,\mathbf{q}}}{\partial q_\gamma} \right\rangle \tag{13}$$

Finally, this can be reduced, using Equation (5) and the orthogonality relation $\sum_{\mathbf{R}} e^{i(\mathbf{q}'-\mathbf{q})\cdot\mathbf{R}} = (2\pi)^3\delta(\mathbf{q}'-\mathbf{q})/\Omega$, to obtain the desired expression for the macroscopic magnetization [Eq. (14)]:

$$M_{0,\alpha} = -\frac{i\varepsilon_{\alpha\beta\gamma}}{2c(2\pi)^3} \sum_n \int_{\mathrm{BZ}} \mathrm{d}\mathbf{q} \left\langle \frac{\partial u_{n,\mathbf{q}}}{\partial q_\beta} \middle| H(\mathbf{q}) \middle| \frac{\partial u_{n,\mathbf{q}}}{\partial q_\gamma} \right\rangle \tag{14}$$

This is our principal result. An identical expression can be obtained by working in the crystal-momentum representation, without any reference to the WFs. We also notice that the integrand in Equation (14) is similar, though not identical, to the orbital magnetization of a wavepacket, as obtained by Sundaram and Niu[20] within the semiclassical approximation.

This equation may be rewritten in a manifestly gauge-invariant form as Equation (15),

$$M_{0,\alpha} = -\frac{i\varepsilon_{\alpha\beta\gamma}}{2c(2\pi)^3}\sum_n \int_{\mathrm{BZ}} \mathrm{d}\mathbf{q}\langle\frac{\partial u_{n,\mathbf{q}}}{\partial q_\beta}|Q_{n,\mathbf{q}}H(\mathbf{q})Q_{n,\mathbf{q}}|\frac{\partial u_{n,\mathbf{q}}}{\partial q_\gamma}\rangle \qquad (15)$$

where $Q_{n,\mathbf{q}}=1-|u_{n,\mathbf{q}}\rangle\langle u_{n,\mathbf{q}}|$. Here, we have made use of the antisymmetrization in Equation (14) and the fact that the quantity $\langle u_{n,\mathbf{q}}|\partial/\partial q_\alpha|u_{n,\mathbf{q}}\rangle$ is pure imaginary. Now, $Q_{n,\mathbf{q}}(\partial/\partial q_\alpha)|u_{n,\mathbf{q}}\rangle$ is manifestly gauge-covariant, in the sense that $Q_{n,\mathbf{q}}(\partial/\partial q_\alpha)[e^{i\beta(\mathbf{q})}|u_{n,\mathbf{q}}\rangle]=e^{i\beta(\mathbf{q})}[Q_{n,\mathbf{q}}(\partial/\partial q_\alpha)|u_{n,\mathbf{q}}\rangle]$ for arbitrary phase twist $\beta(\mathbf{q})$, as is easily checked. (Indeed, $Q_{n,\mathbf{q}}(\partial/\partial q_\alpha)$ is often referred to as the "covariant derivative" for this reason[21]). Thus, Equation (15) is gauge-invariant in a strong sense: not only the integral, but even the integrand, is gauge-invariant. (Actually, this is also true of Equation (14), though less obviously so.)

Note that Equation (15) is invariant with respect to a change of zero of the Hamiltonian. Such a shift gives rise to a contribution proportional to the integral of Equation (16):

$$\Omega_{\alpha\beta} = i\sum_n \langle\frac{\partial u_{n,\mathbf{q}}}{\partial q_\beta}|Q_{n,\mathbf{q}}|\frac{\partial u_{n,\mathbf{q}}}{\partial q_\gamma}\rangle \qquad (16)$$

over the BZ. The quantity in Equation (16) is known as the "Berry curvature," and its BZ integral is just the Chern number.[12,13] Since we restricted ourselves to the case of conventional insulators having zero Chern number (see Section 2), this integral vanishes. Also, note that while Equation (14) and (15) remain well defined and gauge invariant even for a metal, their use in the metallic case is not justified according to the approach followed here.

4. Discretization

We discretize Equation (15) on a simple cubic mesh in reciprocal space, whose grid unit vectors are $\boldsymbol{\kappa}_\alpha=\Delta\mathbf{e}_\alpha$, where $\mathbf{e}_\alpha$ are unit vectors along the Cartesian axes. On this grid, the covariant derivative can be approximated as Equation (17),[22]

$$Q_{n,\mathbf{q}}|\frac{\partial u_{n,\mathbf{q}}}{\partial q_\alpha}\rangle = \frac{1}{2\Delta}(|\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_\alpha}\rangle-|\tilde{u}_{n,\mathbf{q}-\boldsymbol{\kappa}_\alpha}\rangle) \qquad (17)$$

where Equation (18) is valid,

$$|\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_\alpha}\rangle = \frac{|u_{n,\mathbf{q}+\boldsymbol{\kappa}_\alpha}\rangle}{\langle u_{n,\mathbf{q}}|u_{n,\mathbf{q}+\boldsymbol{\kappa}_\alpha}\rangle} \qquad (18)$$

since the inner product of $\langle u_{n,\mathbf{q}}|$ with the right-hand side of Equation (17) then vanishes exactly as it should. The z component of the magnetization can then be written as Equation (19),

$$M_{0,z} = \frac{-\Delta}{32\pi^3 c}\sum_{n,\mathbf{q}}[C^{(++)}_{n,\mathbf{q}}+C^{(-+)}_{n,\mathbf{q}}+C^{(+-)}_{n,\mathbf{q}}+C^{(--)}_{n,\mathbf{q}}] \qquad (19)$$

where the $C^{ACHTUNGTRENNUNG(\mu\nu)}_{n,\mathbf{q}}$ are defined in Equations (20)–(23):

$$C^{(++)}_{n,\mathbf{q}} = \mathrm{Im}\langle\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_y}|H(\mathbf{q})|\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_x}\rangle \qquad (20)$$

$$C^{(-+)}_{n,\mathbf{q}} = \mathrm{Im}\langle\tilde{u}_{n,\mathbf{q}-\boldsymbol{\kappa}_x}|H(\mathbf{q})|\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_y}\rangle \qquad (21)$$

$$C^{(--)}_{n,\mathbf{q}} = \mathrm{Im}\langle\tilde{u}_{n,\mathbf{q}-\boldsymbol{\kappa}_y}|H(\mathbf{q})|\tilde{u}_{n,\mathbf{q}-\boldsymbol{\kappa}_x}\rangle \qquad (22)$$

$$C^{(+-)}_{n,\mathbf{q}} = \mathrm{Im}\langle\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_x}|H(\mathbf{q})|\tilde{u}_{n,\mathbf{q}-\boldsymbol{\kappa}_y}\rangle \qquad (23)$$

Intuitively, for each q_z of the grid, the (q_x,q_y) plane is tessellated into triangular plaquettes, each giving a contribution $C^{(\mu\nu)}_{n,\mathbf{q}}$ as given in Equation (20–23). (Actually, the tessellation covers the (q_x,q_y) plane twice, since there are overlapping triangles arising from neighboring k-points.) Expressions for $M_{0,x}$ and $M_{0,y}$ are obtained by an obvious correspondence.

It is straightforward to check that Equation (19) is invariant with respect to a shift of H by a constant energy E_0. The extra contribution at $\mathbf{q}$ is proportional to the sum over four terms like $\mathrm{Im}\langle\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_y}|\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_x}\rangle$. Since each of these is of the form $\mathrm{Im}[1+\mathcal{O}(\Delta^2)]$ it is permissible, to $\mathcal{O}(\Delta^2)$, to replace $\mathrm{Im}\langle\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_y}|\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_x}\rangle$ by $\mathrm{Im}\ln\langle\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_y}|\tilde{u}_{n,\mathbf{q}+\boldsymbol{\kappa}_x}\rangle$. Then, the sum of these four terms becomes precisely the Berry phase around the diamond-shaped plaquette surrounding point $\mathbf{q}$. Summing over all $\mathbf{q}$, we get a contribution that is proportional to the total Berry phase obtained by circulating around the boundaries of the entire 2D BZ, which vanishes in a periodic gauge.

5. Numerical Tests

We have checked the correctness of our analytic formula, Equation (15), by numerical calculations on the 2D Haldane model.[12] This model is comprised of a honeycomb lattice with two tight-binding sites per cell with site energies $\pm E_0$, real first-neighbor hoppings t_1, and complex second-neighbor hoppings $t_2e^{\pm i\phi}$ as in Figure 1 of ref. [12]. This is a model in which there is no macroscopic magnetic field, although it can be regarded as having been constructed by threading flux tubes of opposite signs through different portions of the unit cell. For our tests, we have chosen $E_0=2$, $t_1=1$ and $t_2=1/3$ and allowed ϕ to vary. (With this choice, states of nonzero Chern number are avoided.[12]) We treat the lower band as occupied and the upper band as empty.

We have computed several quantities and compared them as shown in Table 1. The second column shows the result of a bulk calculation in which we computed the Bloch functions on a 300×300 k-space mesh and computed M_0 using the approach of Equation (19).[23] The remaining columns show results drawn from calculations on large but finite rhombus-shaped samples of size $N\times N$ (that is, containing N^2 sites of energy $-E_0$ and $+E_0$ each). The N^2 occupied WFs were obtained by acting with the band projector on a set of N^2 trial functions (here, just delta functions on the $-E_0$ sites) and applying a subsequent symmetric orthonormalization. The quantity M_{central} in the third column of Table 1 represents the circulation calculated for a single bulklike WF selected from the center of the finite sample. It can be seen that M_0 and M_{central} are essentially identical, thus validating the formal derivations given in the earlier sections.

Table 1. Numerical calculations of various contributions to the orbital magnetization for several values of the flux parameter ϕ in the Haldane model.

ϕ	M_0	$M_{central}$	M_{macro}	M_{edge}	M_0+M_{edge}
$\pi/4$	0.01741	0.01741	0.00512	−0.01229	0.00512
$\pi/2$	0.02835	0.02835	0.00000	−0.02835	0.00000
$3\pi/4$	0.01229	0.01229	−0.00512	−0.01741	−0.00512

We then computed the *total* circulation of the entire $N\times N$ finite sample and divided by the area to get the macroscopic average quantity reported as M_{macro} in the fourth column. (The results shown are extrapolated to $N\rightarrow\infty$ from calculations for $N=10$, 20, and 30). Clearly M_{macro} should correspond to the physical orbital magnetization, but the result *is not* identical to the ones in the previous columns.

In tracing the source of this discrepancy, we found that, while the WFs in the center of the finite sample carry no net current, the WFs at the edge of the sample *do generally carry* nonzero current (i.e. $\langle w_j|\mathbf{v}|w_j\rangle\neq 0$). Summing the tangential components of the net currents associated with the last five WFs at the right edge of the sample gives the values reported as M_{edge}. As shown in the last column of Table 1, it is clear that $M_0+M_{edge}=M_{macro}$ to the precision of the calculation. (The antisymmetry of M_{macro} about $\phi=\pi/2$ can be traced to a particle–hole symmetry of the Haldane model.[24])

Thus, it appears that we have identified all of the contributions that need to be included in a theory of the orbital magnetization. However, it remains unclear whether M_{edge} is a bulk property, and if so, how to compute it from a knowledge of the bulk Bloch functions alone.

6. Summary and Conclusions

We have presented a formalism for the calculation of an important contribution to the orbital magnetization in extended systems with broken time-reversal symmetry. We have restricted ourselves to insulators in vanishing macroscopic magnetic fields, and have considered only the case of isolated bands of zero Chern number.[25] Our approach utilizes WFs to resolve the ill-defined character of the circulation operator in periodic systems, although an identical result can be derived using the crystal-momentum representation to work directly with Bloch orbitals. These considerations lead to an expression for a magnetization contribution $\mathbf{M}_0$ which involves a BZ integral over the Hamiltonian sandwiched between covariant derivatives, Equation (14). This expression is manifestly gauge-invariant and can easily be calculated using the discretization in reciprocal space given in Equation (19).

Our numerical tests confirm that Equation (19) is correct, but they also indicate that $\mathbf{M}_0$ is not the only contribution to the full orbital magnetization $\mathbf{M}$. Evidently it is also necessary to include a second contribution that manifests itself, in the Wannier representation, in terms of extra net currents flowing on WFs near the surface. Our work leaves open the question whether this extra contribution can also be expressed as a bulk property, computable from a knowledge of the bulk Bloch functions. This is clearly an important direction for future research.

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[23] A factor of $\Delta/2\pi$ is removed from Equation (19) for the two-dimensional case.
[24] Each contribution to M is 2π-periodic and odd under $\phi\rightarrow-\phi$ (time reversal). In addition, M_{macro} is invariant under $\phi\rightarrow\phi+\pi$. This follows because $\phi\rightarrow\phi+\pi$ is essentially equivalent to $H\rightarrow-H$ (a rotation by 60° and a sign reversal of basis functions on one sublattice must be inserted to relate them, but these operations do not affect the magnetization). The assertion can then be proven by observing that $H\rightarrow-H$ reverses all currents and also interchanges the identity of the top and bottom bands and by using the fact that the M_{macro} of the two bands must sum to zero.
[25] Formally, the theory applies also to the case of a band of zero Chern number when a large, "commensurate" macroscopic magnetic field is present (i.e. when there is an integer number of flux quanta per cell).

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Orbital Magnetization in Extended Systems

A reciprocal-space expression for the bulk magnetization of a crystalline insulator is obtained, and a version suitable for practical implementation in discretized reciprocal space is also derived. Tests on a tight-binding model indicate the presence of additional edge currents. These calculations might be the first step in the direction of a more general theory.

